Evanescent-wave acceleration of ultrashort electron pulses

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High-power femtosecond laser pulses have been used to excite surface plasmons in 500 Å silver and gold films. Nonlinear excitation results in the emission of electron bunches through multiphoton excitation at low power and laser-induced field emission at high power. The energies of photoelectrons are found to extend as high as 0.4 keV. Calculations show that these high energies are due to ponderomotive acceleration in an evanescent field extending from the metal film out into the vacuum. The theoretical calculations suggest that femtosecond electron pulses with relativistic energies can be generated using longer wavelengths or by developing the surface morphology. [DOI: 10.1063/1.1406562]

A number of schemes based on plasma waves and wake fields have been developed to accelerate electrons using high-power lasers with the objective of building the next generation of compact high-gradient accelerators.[1] Because laser light is a transverse electromagnetic wave, the acceleration is also transverse unless complicated schemes are devised to produce a light gradient field. Here, we will present a scheme in which electrons are accelerated by an evanescent electromagnetic field. In the experiments, electron bunches are generated through the femtosecond laser excitation of surface plasmons (SPs) in a thin metal film deposited on one side of a glass prism. At 50 MW/cm^2 incident laser intensity or below, the electron-generation process is dominated by the multiphoton photoelectric effect.[2] At higher incident intensities, however, a strong high-frequency evanescent field ($\sim 10^{10}$ V/cm, 375 THz) is created on the metal surface resulting in a ponderomotive potential. This causes thinning of the potential barrier for electrons escaping from the metal thus increasing the probability of laser-induced field emission and resulting in the charged particles being pushed away from the surface. A maximum kinetic energy of ~0.4 keV gained from the evanescent wave is observed in the experiments. However, it is found that there are several potential methods that could increase electron energy and production efficiency.

In the experiments, two different laser systems operating in different intensity regimes have been used. For medium power experiments, a 250 kHz repetition-rate Ti:sapphire-based regenerative amplifier has been used, producing 3 μJ pulses. For high-power experiments, a 1 kHz regenerative amplifier producing 0.8 mJ pulses has been used. In all cases, the laser pulse width is 100–150 fs and the center wavelength ~800 nm. Approximately 500 Å thick films of gold and silver were vacuum deposited (at 10^{-5} Torr and afterwards exposed to air for short periods) on the hypotenuse face of a 15 mm side-length BK7 right-angle prism. Afterwards, the metal surfaces were examined with an atomic force microscope. The laser beam entered the prism through one of the faces opposite the hypotenuse in order to excite SPs in the metal film in Kretschmann geometry.[3] The experiments were performed in a vacuum chamber at a pressure of 10^{-6}–10^{-7} Torr. The prism was mounted on the outside of the vacuum chamber with the metal film directed towards the interior, permitting the electrons to travel through a hole to the detection system. Maximum electron emission was observed for transverse magnetic (TM) polarization and an internal angle of incidence of 41.3°/42.4° in silver/gold consistent with the known refractive indices of BK7 and the metals at 800 nm, and SP excitation. No electron emission was detectable for angles away from the SP-resonance angle or for transverse electric (TE)-polarized laser light. Special attention had to be paid to avoid laser ablation of the films and white-light continuum generation in the prism above 10 TW/cm^2 incident laser intensity. This incident intensity corresponds approximately to the damage threshold of the material for this pulse duration and experimental geometry.[4] The laser intensity was adjusted using a half-wave retardation plate and a polarizer. To measure the total photocurrent, copper collector plates were used as electron detectors with the potential difference between the prism and the collector typically set between zero and a few tens of volts. The current flowing from the collector to the prism is measured using lock-in detection and the energy distribution of the electrons is measured with a time-of-flight (TOF) spectrometer consisting of a multichannel array plate (MCP) placed 32 cm from the electron source and a 1 GHz bandwidth oscilloscope for data acquisition.

Figure 1 shows the total photoelectron current measured as a function of incident laser power. At zero bias voltage between the prism and collector, the current increases approximately as the fourth power of the incident laser intensity before saturating above 30 GW/cm^2. This is consistent with the third-order dependence that has been reported previously for gold surfaces excited with low-energy 625 nm laser pulses.[2,3,5] When a reverse bias is applied, the current is reduced and the power dependence becomes of higher order, inconsistent with a simple multiphoton-excitation process.[6] The angular distributions of the photoelectrons in the plane of incidence of the laser and perpendicular to it were measured with an array detector consisting of 10 copper collector strips. The in-plane distribution was found to be directional with the majority of photoelectrons emitted perpendicular to

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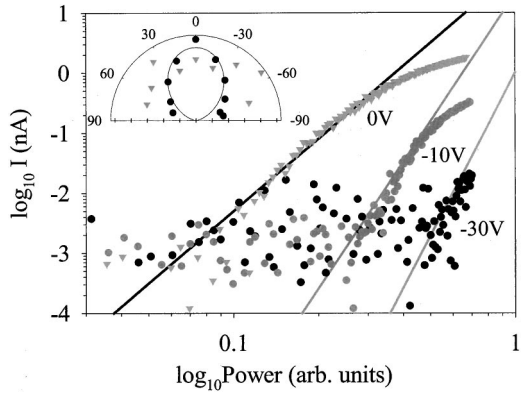

FIG. 1. Dependence of the measured photocurrent from gold on incident laser power. The data were measured with dc bias between the prism and the detector as indicated and with incident power density ranging from ~5 to ~50 GW/cm^2. The lines show fourth-, seventh- and ninth-order power dependencies. The inset shows the measured angular distribution of photoelectrons in the plane of incidence (dots) and perpendicular to that (triangles), as well as a fit to a $\cos^2\vartheta$ distribution.

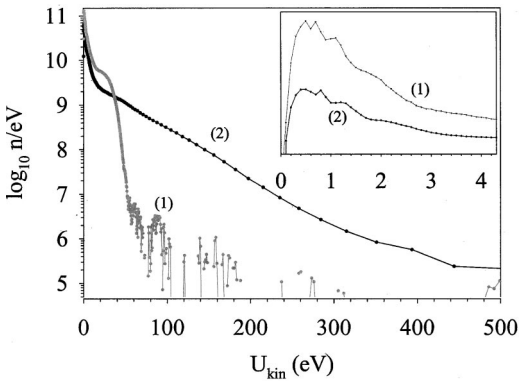

FIG. 2. Kinetic-energy distribution of photoelectrons from a silver film measured as a function of incident intensity. The inset shows the peak of electrons of low kinetic energies on a linear scale. The incident power densities are 12.5 GW/cm^2 (1) and 40 TW/cm^2 (2). The experimental data were averaged into 0.1 eV energy bins, resulting in an energy resolution ranging from 0.1 eV at low energy to 40 eV at 0.4 keV.

the metal surface (Fig. 1). It fits reasonably well to a $\cos^2\vartheta$ function and is independent of the applied bias (within the signal-to-noise ratio). In contrast, the out-of-plane distribution is much more uniform and a preferred direction of photoelectron emission cannot be distinguished. Because of the boundary conditions, the SP electric field must be perpendicular to the film and its wave vector in the plane of the film. When SPs are formed, they will acquire momentum directed into the plane of the film. Thus, one would expect the electrons to travel through the film, rather than come out of it. In fact, it has been observed in the low incident laser-intensity regime (<0.2 GW/cm^2) that SPs have a 13 μm mean free path in the metal film.[7]

There are numerous studies of the lifetimes of electrons and plasmons which show that in metals the *energy* relaxation time is one the order of a few picoseconds and the *momentum* relaxation time is ~40 fs (or less at higher energies).[8–10] Therefore, the momentum of excited electrons should be randomized within a fraction of a laser-pulse duration while the average energy remains constant. Thus, once excited above the Fermi level, the electrons should emerge from the film with a randomized orientational distribution in all directions. This, however, is inconsistent with the observed directionality of the electron beam emitted. We conclude that another process must be responsible for pushing the electrons away from the surface.

Figure 2 shows the measured kinetic-energy distribution of photoelectrons emitted from a silver film. The total current of emitted electrons is about 10 nA at a 250 kHz repetition rate corresponding to 40 fC per bunch. For 0.5 GW/cm^2 incident power density it is found that the distribution has a half width at half maximum of ~0.2 eV. The reason for this narrow width (compared to what might be expected from a Fermi–Dirac distribution) is that the SP resonance selects electrons with a narrow range of momenta. With increasing laser power density, the peak broadens, with a maximum at about 0.5 eV, and there appears to be a cutoff energy. This cutoff energy is proportional to the incident laser power and is about 50 eV above the work function of the metal (corresponding to 33 photons) for 16.1 GW/cm^2. In principle, it is possible to produce electrons with very high kinetic energies by increasing the incident laser power (Fig. 2, data set 2). In that case, however, the damage threshold of the sample can be exceeded, resulting in its degradation. To obtain data (1) in Fig. 2 the incident beam was collimated with a 6 mm diameter. Only about 15% of incident light was absorbed by the metal layer suggesting that further improvements in electron yield and energy are possible. The high kinetic energies measured (as high as 0.4 keV) are not due to multiphoton absorption because in that case one would expect to observe a series of peaks in the spectrum separated by 1.5 eV. Such peaks have not been observed for incident power densities ranging from 0.5 GW/cm^2 to 40 TW/cm^2. However it is possible that nonlinearity in the film causes optical rectification of the incoming laser beam, resulting in an accelerating dc field, which can generate a continuous spread of electron energies. Such an effect was indeed studied but the electric field was expected to be extremely weak.[10]

If the field strength on the prism surface is $E_0(t)$, the field as a function of position z away from the surface is $E(z,t)=E_0(t)\cos(\omega t)\exp(-z/\lambda)$ where λ is the evanescent-wave decay length calculated to be ~1 μm for 800 nm wavelength. The Lorentz force on the electrons results in the nonlinear equation of motion $\ddot{z}=f\cos(\omega t)\exp(-z/\lambda)$, where $f\equiv eE_0/m$, e is the electron charge, m the electron mass, and ω the angular frequency of the laser field. The electron motion consists of fast and slow drifts away from the high field region. Solving it as a classical equation of averaged motion[11] one can express the energy gain of an electron as $U=U_{\text{final}}e^{-2z/\lambda}$, $U_{\text{final}}=e^2E_0^2/(4m\omega^2)$ were U is the ponderomotive potential. This results in an effective dc electric field, which on the surface has magnitude $E_{\text{effective}}=eE_0^2/(2\lambda m\omega^2)$. Using our laser parameters (~0.8 mJ/pulse, 100 fs, beam waist 100 μm) one calculates a field strength of ~3 10^8 V/cm. It is also calculated that excitation of SPs in flat silver or gold films on BK7 results in field enhancement of about a factor of 10. In addition, local nonuniformities on the metal surface are expected to enhance the electric field further.[12] After detailed examination of the metal films used in the experiments features approximately 10 nm high and 20 nm wide were revealed, suggesting that

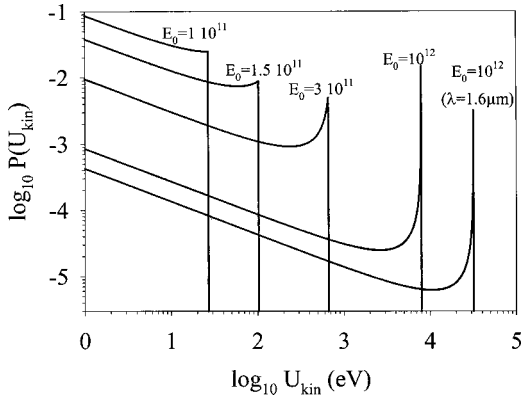

FIG. 3. Theoretical probability-distribution functions of photoemitted electrons. The calculation was performed with zero initial kinetic energy, $\lambda = 800$ nm, $\tau_{pulse} = 100$ fs and the surface field strength indicated in V/m.

the overall field enhancement might be magnified by the localization of SPs. Therefore the overall electric field on the surface of the metal is expected to exceed $\sim 3 \times 10^9$ V/cm.

When an electron is ejected from the metal layer, the evanescent optical field pushes it away from the surface and it would gain energy U_{final} if the field remained on infinitely long. To calculate an energy-distribution function for the photoelectrons, an analytical solution to the equation of motion was used while assuming a 100 fs square laser pulse and constant photoelectron production during the pulse. Thus, electrons created at the beginning of the pulse will be accelerated for the entire duration of the pulse, whereas electrons created at the end of the pulse will not be accelerated at all, resulting in a distribution of energies. Figure 3 shows the calculated kinetic-energy distributions for a series of surface electric-field strengths relevant to our experiments. These distributions reproduce the observed cutoff energy although the unrealistic assumption of a square pulse results in a much sharper cutoff than that measured in the experiment. For low laser intensity, the cutoff energy scales quadratically with the laser intensity. If the laser intensity or the pulse duration is increased, the kinetic energy will saturate and the final kinetic energy will scale as U_{final}.

The experiments and theory presented here show that evanescent fields produced by amplified femtosecond laser pulses can be used very effectively to accelerate electrons. In our experiments, we are limited to peak power densities of ~ 40 TW/cm^2 and a wavelength of 800 nm. Using the theory, one can extrapolate the results to other regimes. It should be possible to obtain much higher energies by using infrared femtosecond laser pulses or by changing the surface morphology. This could even lead to the generation of electrons with relativistic energies.

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